

UV CURABLE COMPOSITIONS FOR PRODUCING
ELECTROLUMINESCENT COATINGS

Instal

TECHNICAL FIELD

5 The present invention relates to the active layer in an electroluminescent device.

BACKGROUND ART

10 A typical electroluminescent device is a multilayer thin film structure that emits visible light when activated by an applied voltage. The active layer in such a device will contains a phosphor. This active layer has previously been deposited applying various curable compositions to a suitable substrate followed by ultraviolet (UV) light curing or heat curing. The usual compositions, however, contain organic solvents that do not incorporate into the active layer after curing. Such solvent based systems are undesirable because of the hazards and expenses associated with volatile organic solvents.

15 UV radiation curable compositions are applied to a substrate through spraying, screen printing, dipping or brushing for the protection or decoration of the substrate. In the usual application, a substrate such as metals, glass, or plastics is coated with the composition and then UV light is introduced to compete the curing process. The UV curable compositions offer many advantages over typical heat
20 curable compositions.

Heat curable compositions require the use of organic solvents that contain a significant amount of volatile organic compounds (VOCs). These VOCs escape into the atmosphere while the heat curable composition dries. Such solvent based systems are undesirable because of the hazards and expenses associated with
25 VOCs. The hazards include water and air pollution and the expenses include the cost of complying with strict government regulation on solvent emission levels. In

contrast, UV curable compositions contain reactive monomers instead of solvents; thus eliminating the detrimental effects of the VOCs.

5 The use of heat curable compositions not only raises environmental concerns but other disadvantages exist with their use as well. Heat curable compositions suffer from slow cure times which lead to decreased productivity. These compositions require high energy for curing due to energy loss as well as the energy required to heat the substrate. Additionally, many heat curable compositions yield poor film properties that result in decreased value of the end product.

10 In a typical electroluminescent device, the active layer comprises one layer of a multilayer electroluminescent device. An example of such a device would contain a substrate made of polycarbonate or glass coated with a transparent conductor such as fluorine doped tin oxide. Metallic grid lines are patterned onto the substrate. The active layer is then applied by screen printing the electroluminescent composition onto the substrate with gridlines. A dielectric
15 coating is then optionally applied over the structure. Finally, the device is coated with a metallic backing. The active layer is such electroluminescent devices typically contains a phosphor. Such phosphor may or may not be encapsulated with various oxides or nitrides. Encapsulation protects the phosphor from the deleterious environmental effects.

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DISCLOSURE OF INVENTION

It is an object of the present invention to provide an improved electroluminescent composition that is curable by ultraviolet light.

It is another object of the present invention to provide an improved electroluminescent composition that can be applied by spraying, screen printing,
25 dipping, and brushing.

It is still another object of the present invention to provide an improved electroluminescent composition that comprises either an encapsulated or an unencapsulated phosphor.

5 It is yet another object of the present invention to provide an improved electroluminescent composition that comprises at least one aliphatic acrylated oligomer that can be used to coat a substrate such that no significant amount of volatile organic solvents do not become incorporated in the coating after the composition is cured.

10 The present invention discloses an ultraviolet light curable electroluminescent composition and method for making such a composition that may be used to produce an electroluminescent active layer. In this context, an active layer is a layer that when incorporated in a suitable device emits light when a voltage is applied. The disclosed composition does not contain any significant amount of volatile organic solvents that do not become incorporated in the active layer after
15 curing. Specifically, the electroluminescent composition contains 5% or less volatile organic solvents by weight. It is an advantage of the present invention that the deposition of the dielectric layer in such a device is optional when the disclosed electroluminescent composition is used to deposit the active layer.

20 In accordance with one aspect of the invention, an ultraviolet light curable electroluminescent composition is provided. The electroluminescent composition comprises a mixture of one or more aliphatic acrylated oligomers, wherein the aliphatic acrylated oligomer mixture is present in an amount of about 10% to 40% of the electroluminescent composition. All percentages of the electroluminescent composition as expressed in this document refer to the weight
25 percentage of the stated component to the total mass of the electroluminescent composition.

The electroluminescent composition preferably comprises an isobornyl acrylate monomer in an amount of about 4% to 30% of the electroluminescent composition, optionally an adhesion promoter in an amount of

1% to 10%, a photoinitiator in an amount of about 0.5% to 6% of the electroluminescent composition, optionally, a flow promoting agent in an amount of about 0.1% to 5% of the electroluminescent composition, and an electroluminescent phosphor in an amount of 28% to 80%. The electroluminescent phosphor may either be encapsulated or unencapsulated.

In accordance with yet another aspect of the invention, a method is provided for depositing a electroluminescent coating on a substrate. The method comprises a first step of applying to the substrate a electroluminescent-containing fluid-phase composition ("electroluminescent composition"). The electroluminescent composition comprises a mixture of aliphatic acrylated oligomers, wherein the aliphatic acrylated oligomer is present in an amount of about 10% to 40% of the electroluminescent composition. The electroluminescent composition also includes an isobornyl acrylate monomer in an amount of about 4% to 30% of the electroluminescent composition, a photoinitiator in an amount of about 0.5% to 6% of the electroluminescent composition, a flow promoting agent in an amount of about 0.1% to 5% of the electroluminescent composition, a copper activated zinc sulfide electroluminescent phosphor in an amount of 28% to 80%, and optionally an adhesion promoter in an amount of 1% to 10%.

The method also includes a second step of illuminating the electroluminescent composition on the substrate with an ultraviolet light to cause the electroluminescent composition to cure into the electroluminescent coating.

In accordance with this method, the electroluminescent composition can be selectively deposited on the substrate at specific locations where electroluminescent plating is desired. It need not be applied to the entire substrate.

BRIEF DESCRIPTION OF THE DRAWINGS

Figure 1 is a graph that the light intensity over time of an electroluminescent device incorporating an active layer made with the composition of the present invention.

BEST MODE FOR CARRYING OUT THE INVENTION

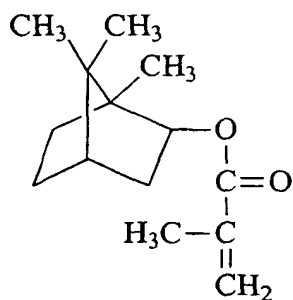
Electroluminescent Compositions

Reference will now be made in detail to presently preferred compositions or embodiments and methods of the invention, which constitute the best modes of practicing the invention presently known to the inventor.

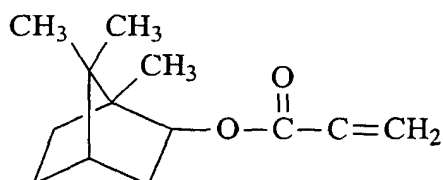
In accordance with one aspect of the invention, a presently preferred ultraviolet light curable electroluminescent composition ("electroluminescent composition") is provided. In this preferred embodiment, the electroluminescent composition includes a mixture of aliphatic acrylated oligomers. The aliphatic acrylated oligomer mixture is preferably present in an amount of about 10% to 40% of the weight of the electroluminescent composition. In a particularly preferred embodiment the aliphatic acrylated oligomer mixture is present in an amount of about 34% of the weight of the electroluminescent composition. In another particularly preferred embodiment the aliphatic acrylated oligomer mixture is present in an amount of about 12% of the weight of the electroluminescent composition. The aliphatic acrylated oligomer preferably comprises one or more urethane oligomers. Suitable aliphatic acrylated oligomers include Radcure Ebecryl 244 (aliphatic urethane diacrylate diluted 10% with 1,6-hexanediol diacrylate), Ebecryl 264 (aliphatic urethane triacrylate diluted 15% with 1,6-hexanediol diacrylate), Ebecryl 284 (aliphatic urethane diacrylate diluted 10% with 1,6-hexanediol diacrylate) commercially available from Radcure UCB Corp. of Smyrna, Georgia; Sartomer CN-961E75 (aliphatic urethane diacrylate blended with 25% ethoxylated trimethylol propane triacrylate), CN-961H81 (aliphatic urethane diacrylate blended with 19% 2(2-ethoxyethoxy)ethyl acrylate), CN-963A80 (aliphatic urethane diacrylate blended with 20% tripropylene glycol diacrylate), CN-964 (aliphatic urethane diacrylate), CN-966A80 (aliphatic urethane diacrylate blended with 20% tripropylene glycol diacrylate), CN-982A75 (aliphatic urethane diacrylate blended with 25% tripropylene glycol diacrylate) and CN-983 (aliphatic urethane diacrylate), commercially available from Sartomer Corp. of Exton, Pennsylvania; TAB FAIRAD 8010, 8179, 8205, 8210, 8216, 8264, M-E-15, UVU-316, commercially available

from TAB Chemicals of Chicago, Illinois; and Echo Resin ALU-303, commercially available from Echo Resins of Versailles, Missouri; and Genomer 4652, commercially available from Rahn Radiation Curing of Aurora, IL. The preferred aliphatic acrylated oligomers include Ebecryl 264 and Ebecryl 284. Ebecryl 264 is an aliphatic urethane triacrylate of 1200 molecular weight supplied as an 85% solution in hexanediol diacrylate. Ebecryl 284 is aliphatic urethane diacrylate of 1200 molecular weight diluted 10% with 1,6-hexanediol diacrylate. Combinations of these materials may also be employed herein.

The preferred electroluminescent composition also includes an isobornyl acrylate monomer preferably in an amount of about 4% to 30% of the electroluminescent composition. In one particularly preferred embodiment of the present invention, the isobornyl acrylate monomer is present in an amount of about 20% of the electroluminescent composition. In another particularly preferred embodiment of the present invention, the isobornyl acrylate monomer is present in an amount of about 8% of the electroluminescent composition. Suitable isobornyl acrylate monomers include Sartomer SR423 (isobornyl methacrylate):



and SR506 (isobornyl acrylate):



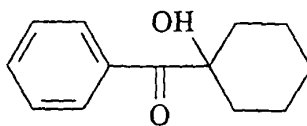
available from Sartomer Corp.; Radcure IBOA (isobornyl acrylate), commercially available from Radcure Corp.; IBOA and IBOMA, commercially available from CPS Chemical of Bradford, England; and Genomer 1121, commercially available from Rahn Radiation Curing. The preferred isobornyl acrylate monomer is Radcure IBOA, commercially available from Radcure Corp. Radcure IBOA is a high purity, low color monomer. Combinations of these materials may also be employed herein.

The preferred electroluminescent composition may also includes an adhesion promoter preferably in an amount of about 1% to 10% the electroluminescent composition. In one particularly preferred embodiment of the present invention the adhesion promoter is present in an amount of about 7%. In another particularly preferred embodiment of the present invention the adhesion promoter is present in an amount of about 3% of the electroluminescent composition. Suitable adhesion promoters include Ebecryl 168, commercially available from Radcure Corp.; and Sartomer CN 704 (acrylated polyester adhesion promoter) and CD 9052 (trifunctional acid ester), commercially available from Sartomer Corp. The preferred adhesion promoter is Ebecryl 168 which is a methacrylated acidic adhesion promoter. Combinations of these materials may also be employed herein.

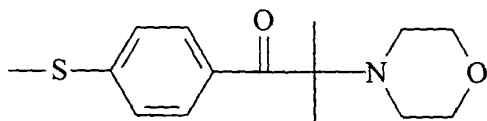
The preferred electroluminescent composition also includes an electroluminescent phosphor. Preferably the electroluminescent phosphor is a copper activated zinc sulfide electroluminescent phosphor. The copper activated zinc sulfide electroluminescent phosphor is preferably present in an amount of about 28% to 80% of the electroluminescent composition. The copper activated zinc sulfide electroluminescent phosphor may either be encapsulated or unencapsulated. In one particularly preferred embodiment of the present composition the zinc sulfide electroluminescent phosphor is present in an amount of about 33%. In another particularly preferred embodiment of the present invention, the electroluminescent phosphor is a mixture of an encapsulated and unencapsulated phosphor present in a total amount of about 80% of the weight of the electroluminescent composition wherein the encapsulated phosphor is about 50% of the electroluminescent composition and the unencapsulated phosphor is about 25% of the electroluminescent composition. Suitable encapsulated electroluminescent phosphors include TNE 100,

TNE 120, TNE 200 TNE 210, TNE 220, TNE 230, TNE 300, TNE 310, TNE 320, TNE 400, TNE 410 TNE 420, TNE 430, TNE 500, TNE 510, TNE 520, TNE 600, TNE 620, TNE 700, TNE 720, NE 100, NE 120, NE 200, NE 210, NE 220, NE 230, NE 300, NE 310, NE 320, NE 400, NE 410 NE 420, NE 430, NE 500, NE 510, NE 520, NE 600, NE 620, NE 700, NE 720, ANE 200, ANE 230, AND 400, and ANE 430 commercially available from Osram Sylvania. Suitable unencapsulated electroluminescent phosphors include 723 EL, 727 EL, 728 EL, 729 EL, 813 EL, and 814 EL commercially available from Osram Sylvania. These materials may emit red, green, yellow, blue or orange colored light based upon the particular phosphor employed. The preferred electroluminescent phosphors are TNE 100, TNE 200, TNE 410, and TNE 700, commercially available from Osram Sylvania.

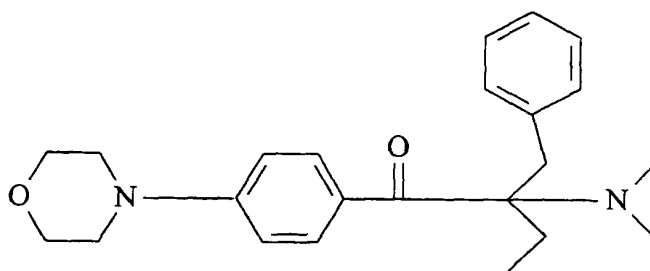
This preferred electroluminescent composition also includes a photoinitiator preferably in an amount of about 0.5% to 6% of the electroluminescent composition. In one particularly preferred embodiment of the present invention the photoinitiator is present in an amount of about 3%. In another particularly preferred embodiment of the present invention the photoinitiator is present in an amount of about 1% of the electroluminescent composition. Suitable photoinitiators include Irgacure 184 (1-hydroxycyclohexyl phenyl ketone),



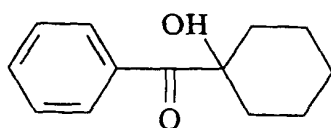
Irgacure 907 (2-methyl-1-[4-(methylthio)phenyl]-2-morpholino propan-1-one),



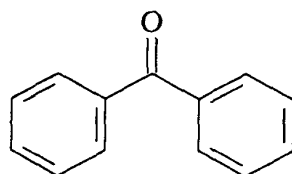
Irgacure 369 (2-benzyl-2-N,N-dimethylamino-1-(4-morpholinophenyl)-1-butanone),



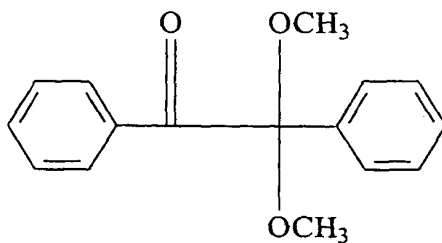
Irgacure 500 (the combination of 50% 1-hydroxy cyclohexyl phenyl ketone,



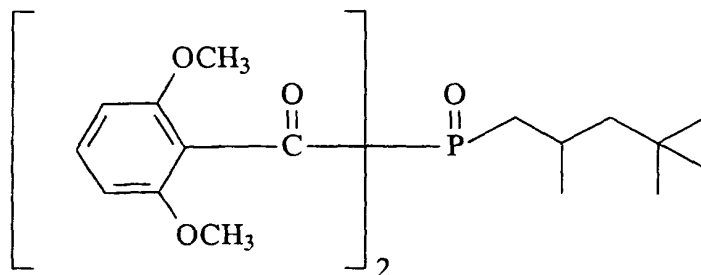
and 50% benzophenone),



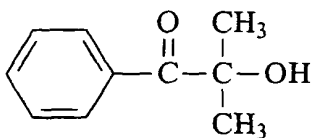
Irgacure 651 (2,2-dimethoxy-1,2-diphenylethan-1-one),



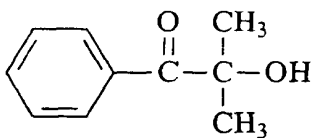
Irgacure 1700 (the combination of 25 % bis(2,6-dimethoxybenzoyl-2,4,4-trimethyl pentyl) phosphine oxide,



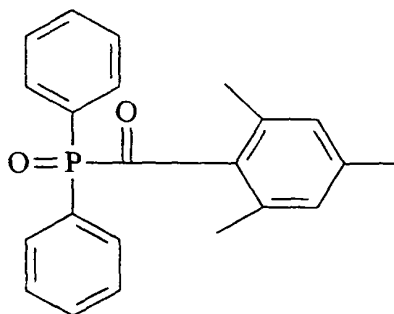
and 75 % 2-hydroxy-2-methyl-1-phenyl-propan-1-one),



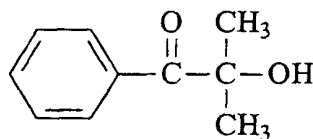
and DAROCUR 1173 (2-hydroxy-2-methyl-1-phenyl-1-propane),



5 and DAROCUR 4265 (the combination of 50% 2,4,6- trimethylbenzoyldiphenyl-phosphine oxide,



and 50 % 2-hydroxy 2-methyl-1-phenyl-propan-1-one),



available commercially from Ciba-Geigy Corp., Tarrytown, N.Y.; CYRACURE UVI-6974 (mixed triaryl sulfonium hexafluoroantimonate salts) and cyracure UVI-6990 (mixed triaryl sulfonium hexafluorophosphate salts) available commercially
5 from Union Carbide Chemicals and Plastics Co. Inc., Danbury, Connecticut; and Genocure CQ, Genocure BOK, and Genocure M.F., commercially available from Rahn Radiation Curing. The preferred photoinitiator is Irgacure 1700 commercially available from Ciba-Geigy of Tarrytown, New York. Combinations of these materials may also be employed herein.

10 The preferred electroluminescent composition still further includes a flow promoting agent preferably in an amount of about 0.1% to 5% of the electroluminescent composition. In one particularly preferred embodiment of the present invention the flow promoting agent is present in an amount of about 3%. In
15 another particularly preferred embodiment of the present invention the flow promoting agent is present in an amount of about 1% of the electroluminescent composition. Suitable flow promoting agents include Genorad 17, commercially available from Rahn Radiation Curing; and Modaflow, commercially available from Monsanto Chemical Co., St. Louis, Missouri. The preferred flow promoting agent is Modaflow which is an ethyl acrylate and 2-ethylhexyl acrylate copolymer that
20 improves the flow of the composition. Combinations of these materials may also be employed herein.

To illustrate, the following example sets forth a presently preferred electroluminescent composition according to this aspect of the invention.

Example 1

This example provides a preferred electroluminescent composition according to the invention that can be used for deposition on the surface of a substrate such as a glass or polycarbonate substrate. Such a substrate may first be coated with a transparent conductor and silver grid lines. The electroluminescent composition was made from the following components:

Component	Approximate Mass %
Ebecryl 264	16.7
Ebecryl 284	16.7
IBOA	20.0
Ebecryl 168	6.7
Modaflow	3.3
Irgacure 1700	3.3
TNE 200	33.3
Total	100.00

In this example the IBOA and Irgacure 1700 are mixed in a pan with a propeller blade mixer for 30 seconds at a speed of 500 to 1000 rpm. In the next step, the Ebecryl 264, the Ebecryl 284, and the Modaflow are introduced into the pan and mixed for 1 to 2 minutes at a speed of 2000 rpm. In the final step, the Ebecryl 168 adhesion promoter and TNE 200 phosphor are introduced into the pan and are mixed for 1 to 2 minutes at a speed of 2000 rpm. The temperature during mixing is monitored. The mixing is temporarily suspended if the temperature exceed 100°F. This particular electroluminescent composition may be used by the method below to produce an active layer that emits bluish-green light.

Example 2

This example provides a preferred electroluminescent composition according to the invention that can be used for deposition on the surface of a

substrate such as a glass or polycarbonate substrate. Such a substrate may first be coated with a transparent conductor and silver grid lines. The electroluminescent composition was made from the following components:

Component	Approximate Mass %
Ebecryl 264	6.3
Ebecryl 284	6.3
IBOA	7.5
Ebecryl 168	2.5
Modaflow	1.3
Irgacure 1700	1.3
ANE 430	49.8
813 EL	25.0
Total	100.00

In this example the IBOA and Irgacure 1700 are mixed in a pan with a propeller blade mixer for 30 seconds at a speed of 500 to 1000 rpm. In the next step, the Ebecryl 264, the Ebecryl 284, and the Modaflow are introduced into the pan and mixed for 1 to 2 minutes at a speed of 2000 rpm. In the final step, the Ebecryl 168 adhesion promoter, ANE 430 phosphor, and 813 EL phosphor are introduced into the pan and are mixed for 1 to 2 minutes at a speed of 2000 rpm. The temperature during mixing is monitored. The mixing is temporarily suspended if the temperature exceed 100°F. This particular electroluminescent composition may be used by the method below to produce an active layer that emits bluish-green light.

Figure 1 is a graph of the change in light output intensity over time for an electroluminescent device incorporating an active layer made from the composition in Example 2. The electroluminescent device is made from a 4 inch by 4 inch glass substrate coated with a transparent conductor. Metallic grid lines are patterned onto the substrate. The active layer is then applied by screen printing the

electroluminescent composition described in Example 2 onto the substrate. Finally, the device is coated with a silver backing. Figure 1 shows that the light intensity of such a device when powered by a 12.5 volt transformer at 100 Hz initially is approximately 20 foot candles per square centimeter and that over a period of approximately two months the light intensity decreases and thereafter stabilizes at a value of approximately 10 foot candles per square centimeter. Furthermore, electroluminescent devices made from the composition in Example 2 have achieved light intensities as high as 88 foot candles per square centimeter for 1 inch by 1 inch devices when powered at 240 watts and 2000 Hz.

10 **Method for Depositing an Electroluminescent Coating on a Substrate**

In accordance with still another aspect of the invention, a method is provided for depositing an electroluminescent coating on a suitable substrate. This method is utilized in depositing the active layer in a multilayer electroluminescent device. The method comprises a first step of applying a phosphor-containing fluid-phase composition ("electroluminescent composition") to the substrate.

The electroluminescent composition comprises an aliphatic acrylated oligomer, the aliphatic acrylated oligomer preferably present in an amount of about 10% to 40% of the electroluminescent composition; an isobornyl acrylate monomer preferably present in an amount of about 4% to 30% of the electroluminescent composition; an photoinitiator preferably present in an amount of a preferably present bout 0.5% to 6% of the electroluminescent composition; a flow promoting agent in an amount of about 0.1% to 5% of the electroluminescent composition, and a zinc sulfide containing phosphor in an amount of 28% to 80%. The preferred electroluminescent compositions according to this method are those described herein, for example, including the compositions described in example 1 and example 2.

The electroluminescent composition may be applied to the substrate using a number of different techniques. The electroluminescent composition may be applied, for example, by direct brush application, or it may be sprayed onto the substrate surface. It also may be applied using a screen printing technique. In such

screen printing technique, a "screen" as the term is used in the screen printing industry is used to regulate the flow of liquid composition onto the substrate surface. The electroluminescent composition typically would be applied to the screen as the latter contacts the substrate. The electroluminescent composition flows through the silk screen to the substrate, whereupon it adheres to the substrate at the desired film thickness. Screen printing techniques suitable for this purpose include known techniques, but wherein the process is adjusted in ways known to persons of ordinary skill in the art to accommodate the viscosity, flowability, and other properties of the liquid-phase composition, the substrate and its surface properties, etc. Flexographic techniques, for example, using pinch rollers to contact the electroluminescent composition with a rolling substrate, also may be used.

The method includes a second step of illuminating the electroluminescent-containing fluid-phase composition on the substrate with an ultraviolet light to cause the electroluminescent-containing fluid-phase composition to cure into the electroluminescent coating. This illumination may be carried out in any number of ways, provided the ultraviolet light or radiation impinges upon the electroluminescent composition so that the electroluminescent composition is caused to polymerize to form the coating, layer, film, etc., and thereby cures.

Curing preferably takes place by free radical polymerization, which is initiated by an ultraviolet radiation source. The photoinitiator preferably comprises a photoinitiator, as described above.

Various ultraviolet light sources may be used, depending on the application. Preferred ultraviolet radiation sources for a number of applications include known ultraviolet lighting equipment with energy intensity settings of, for example, 125 watts, 200 watts, and 300 watts per square inch.

Additional advantages and modifications will readily occur to those skilled in the art. Therefore, the invention in its broader aspects is not limited to the specific details, representative devices, and illustrative examples shown and

described. Accordingly, departures may be made from such details without departing from the spirit or scope of the general inventive concept.

5 While embodiments of the invention have been illustrated and described, it is not intended that these embodiments illustrate and describe all possible forms of the invention. Rather, the words used in the specification are words of description rather than limitation, and it is understood that various changes may be made without departing from the spirit and scope of the invention.